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**OPTICAL PARAMETRIC OSCILLATOR ON GaSe CRYSTAL  
PUMPED BY A 3-MICRON ERBIUM LASER**

**SPONSORED BY  
THE AIR FORCE RESEARCH LABORATORY/PHILLIPS RESEARCH SITE  
THROUGH  
THE EUROPEAN OFFICE OF AEROSPACE RESEARCH AND DEVELOPMENT  
SPC 97-4038  
Contract F61708-97-W0128**

**FINAL REPORT**

**12 May 1998**

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## INTRODUCTION

In the last few years the greater interest has returned to mid-infrared optical parametric oscillators(OPO) pumped by solid state lasers. This was a result of considerable progress in laser and nonlinear crystal growth technologies. A new class of IR solid state lasers based on Ho<sup>3+</sup> and Er<sup>3+</sup>- doped crystals have been developed. Holmium lasers have wavelengths near 2 micron and Erbium lasers near 3 micron. Both these types of lasers can be pumped by flash lamps or laser diodes. The latest laboratory versions of these lasers have efficiencies similar to Nd-doped lasers and Erbium lasers are the longest wavelength high power solid state lasers currently available. These laser sources are very well suited for generating continuously tunable intense and ultrashort IR light pulses using nonlinear optical frequency conversion in optical parametric oscillators based on IR nonlinear crystals. The GaSe crystal has large transparency range(0.65-18 μm) and higher nonlinear figure of merit than other crystals such as CdSe, AgGaSe<sub>2</sub>, which were used for optical parametric oscillators. A lack of a simple and reliable laser with wavelength suited to obtain phase matching condition did not allow, until recently, to obtain parametric generation in this crystal. Lately it was reported on traveling wave parametric oscillation(single pass parametric amplifier without mirrors) in GaSe crystal pumped by single picosecond pulses of YSGG:Cr:Er laser ( $\lambda=2.79 \mu\text{m}$ ). However the highest average power and energy output is obtained in the "classical" OPO. Until now there were no publications on resonator OPO using GaSe crystal. In this project we were trying to fulfill a research program which include investigation of resonator OPO pumped by Q-switched and mode locked erbium laser.

The tuning range of 3-18 μm is of a great interest for spectroscopy because most of the frequencies of molecular vibrations as well as the sub-band transition energies in semiconductor quantum well structures lie in this spectral range. High intensity tunable picosecond laser light could be used for time-resolved saturation spectroscopy in this spectral band. There are several windows of transparency of the atmosphere(the widest is between 3.5-4.2 μm) in the tuning range of such OPOs and these OPO could be used for remote sensing and atmosphere pollutant detection.

Recently, dramatic improvement of cornea ablation by 6.45  $\mu\text{m}$  laser radiation was reported. The 6.45 micron radiation corresponded to the bending vibration mode of water and obtained from free electron laser(FEL). The performance characteristics of OPOs (pulse energy, peak intensity, average power) are comparable with those of a free electron laser. The OPO however is a table top instrument compared with the warehouse sized installation needed to accommodate the electron beam accelerator and magnetic wiggler for a FEL. The equipment and running costs of a properly engineered OPO would be roughly 1/100th of those of the FEL. Aside from the FEL, there are no other tunable laser sources in this range and an OPO-based laser is thus an extremely powerful tool, opening up a whole new region of the spectrum for IR laser applications.

#### TECHNICAL DESCRIPTION

The work on the project consists of several parts. One of them is pumping laser which was used to pump OPO. In the experimental set-up we have used YSGG:Cr:Er ( $\lambda = 2.79 \mu\text{m}$ ) laser crystal which has lower pump energy and higher amplification than YAG:Er( $\lambda = 2.94 \mu\text{m}$ ) laser. YSGG:Cr:Er laser crystals are available from ELMA company (Moscow). For the purposes of nonlinear optical process this small difference in wavelength is not crucial.

An approach that combines high conversion efficiency of a picosecond traveling wave OPO (so called OPG) and high output energy of a resonator OPO is an OPO synchronously pumped by a train of picosecond pulses from a mode-locked laser. Synchronous pump means that the optical length of the OPO cavity is multiple of pump laser cavity. In this optical setup modes in the OPO cavity are locked resulting in low generation threshold and high peak power of tunable IR pulses.

The pumping laser in our project is actively Q-switched and mode-locked Er<sup>3+</sup>:YSGG , generating at 2.79  $\mu\text{m}$ . Some work was made to improve operation of this laser by updating HV electronic driver (output voltage 3-5 kV, pulse duration <100  $\mu\text{s}$ , f=60 MHz) for HF electro-optical modulation of intracavity losses. Output laser pulse in Q-switched mode has output energy up to E=20-22 mJ with pulse duration t~150-200 ns at the repetition rate 1 pps.

Output laser pulse in the mode-locking regime has output energy E~10-15 mJ with pulse duration t~200 ns at the repetition rate 1 pps and with its envelope consisting of 20-30 pulses with single pulse duration 50-100 ps. We were trying to amplify this laser pulse

in an amplifier. However  $\text{Er}^{3+}$  lasers have small one pass gain (1.5-2) but in case of multipass amplification damage of laser crystal may occur.

In experiments on nonlinear OPO we have used GaSe crystals which are available from crystal growth laboratories in Tomsk (Voevodin, Andreev-Gribenukov). Since GaSe crystal is very soft and has layered structure perpendicular to the optical axis, it can not be polished at a certain angle to the optical axis. However, birefringence in this crystal is high enough ( $n_o - n_e = 0.32$ ) to obtain both types of phasematching with 3 micron pump by rotating the crystal at a moderate angle ( $\theta_{int} < 20^\circ$ ). The crystal itself should be put in a special holder. We have studied both type I and II phase matching. With the 3 micron pump at type I (e-oo) phase matching a GaSe OPO generates a wide spectrum IR signal (pseudo-continuum at 4-8  $\mu\text{m}$ ). For type II (e-oe) interaction, the external phase matching angle at the degenerate point is close to the Brewster angle that decreases Fresnel reflection on the faces of the crystal and thus decreases threshold.

Regarding this peculiarity of GaSe we were going to consider different setups of a pump laser and OPO: two different cavities matched by a lens; intracavity OPO in case of pure Q-switched pump; output coupling due to Fresnel reflection; double crystals scheme with rotation of each of them in opposite direction to compensate walk-off of the parametric signal from the pumped area, etc. To obtain tuning in the whole transparency range, different sets of OPO mirrors were needed. Mirrors are available from the 'KVANT' company in Nizhny Novgorod. The needed sets of OPO mirrors was fabricated by the "KVANT".

Unfortunately we couldn't make intracavity OPO because of too high intracavity Fresnel's losses and we couldn't make double crystals scheme because of the one of the two GaSe crystals which we had bought was used for investigations with experimental antireflection coating.

It is extremely important to find a possibility and proper materials for antireflection coating onto GaSe crystal considering its softness and layered structure. Another approach might be the choose of appropriate immerse liquid on GaSe faces. Both approaches are not so simple, taking into account such wide tuning range as, for example, 3-15  $\mu\text{m}$ . But in our opinion without solving this problem wouldn't be possible to make an efficient resonant OPO on GaSe crystal.

We tried to make AR coating onto GaSe crystal consisting of one layer of  $\text{Al}_2\text{O}_3$ , however because of very different mechanical properties of sapphire and GaSe this

coating was not stable and have short lifetime (approximately two weeks). Another material that could be used for simple AR coating for GaSe is LiF. We are planning to try this coating in the future with help of the "KVANT" company. But from the opine of view this contract we couldn't solve this problem.

It was known from the theory of OOPS that the threshold is minimum and conversion efficiency is maximum at the degenerate point where the IR. light frequencies is half of the pump frequency. In our case in the initial experiments tunable IR. light was near 6 micron, but we were trying to achieve tenability in the whole transparency range. It was impossible for us because of too high Fresno losses of uncooked by antireflrcion coating GaSe crystal and hence too high threshold of parametric oscillation comparable or higher then threshold of damage some optical units.

We started with mode-locked pumping because of lower threshold, but OPO conversion efficiency was too small and we had to test resonant GaSe OPO pumped by 3 micron Q-switched laser pulses taking into account that expected conversion efficiency from 3 micron to tunable IR light is of 10-20%.

The minimum topic of interest is the demonstration for the first time resonator OPO on GaSe crystal pumped by a 3 micron laser. However, there is some good and interesting science that can be done in the other areas, such as IR laser architecture and applications of tunable IR light.

In the work was used YSGG:Cr:Er laser at  $\lambda=2.79 \mu\text{m}$  since this laser operates in electrooptic Q-switching and mode locking. Output laser pulse energy is typically  $E=10-15 \text{ mJ}$  in  $\text{TEM}_{00}$ -mode and pulse duration is about  $\tau = 150-200 \text{ ns}$  in Q-switch regime. In the mode-locked regime output energy is approximately the same and  $150-200 \text{ ns}$  envelope consists of 20-30 pulses with single pulse duration 50-100 ps. Repetition rate of the laser equals 1-2 Hz.

For OPO crystal was used GaSe crystal. The Table 1 shows brief comparison of GaSe crystal with well known  $\text{LiNbO}_3$  crystal and some other IR crystals which are used for OPO tasks.

Table 1.

Crystal	$\text{LiNbO}_3$	$\text{Ag}_3\text{AsS}_3$	$\text{CdSe}$	$\text{AgGaSe}_2$	$\text{GaSe}$	$\text{ZnGeP}_2$
Transparency range, $\mu\text{m}$	0.33 - 5.5	0.6 - 13	0.75 - 20	0.71 - 18	0.65 - 18	0.74 - 12
$n_o$ near 3 $\mu\text{m}$	2.16	2.75	2.44	2.65	2.73	3.13
$n_e$	2.09	2.54	2.46	2.62	2.41	3.17
Nonlinearity $d_{\text{eff}}$ ( $10^{-12} \text{ m/V}$ )	5.44	18	18	33	54	88
Figure of merit, $d^2/n^3$ ( $10^{-24} \text{ m}^2/\text{V}^2$ )	3.1	17.6	22	60	128	247

It is seen from the Table that GaSe has good data in comparison with close neighbors  $\text{AgGaSe}_2$  and  $\text{ZnGeP}_2$  (ZGP). GaSe crystal has both advantages and disadvantages.

Advantages of GaSe crystal: wide transparency range without specific points as e.g.  $\text{AgGaSe}_2$  and  $\text{ZnGeP}_2$  at 2 micron; high nonlinearity; lower cost in comparison with ZGP.

Disadvantages of GaSe: very soft- need of special protective holder, but it is not a problem to handle GaSe in a holder; layered structure perpendicular to Z- axis which is optical axis and, therefore, great difficulty in polishing at suitable phasematched angle; however, internal matchangle are relatively small (see matching curve) and the angle could be reach by rotating the crystal without total internal reflection; big birefringence  $n_o - n_e = 0.32$  – need to use wide laser pump beams or/and cylindrical lenses and/or double crystal set-up, this technique are known and well-elaborated.

The calculated tuning curves are presented on the picture. For the calculations were used the data from [K.L.Vodopyanov,L.A.Kulevsky, Optics Communications, 1995,v.118,pp.375-378].

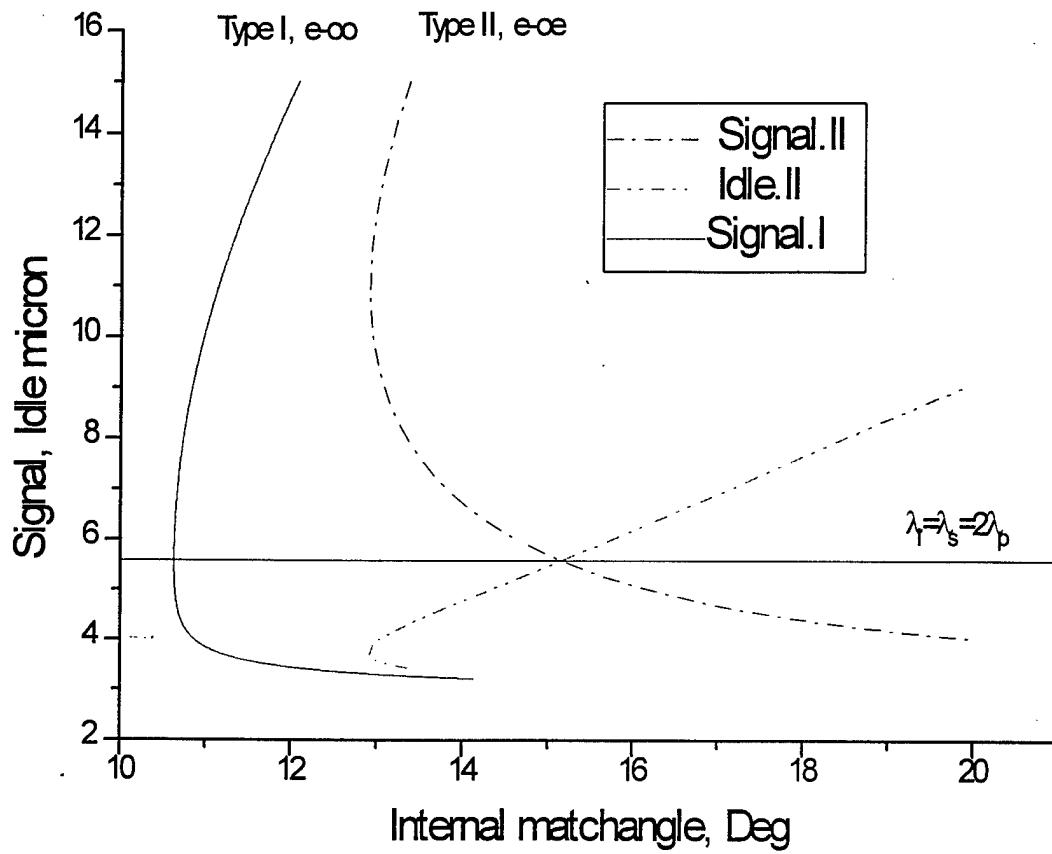


Fig.1

The horizontal solid line near  $6 \mu\text{m}$  corresponds to degeneracy regime of OPO.

Theory gives the increment of single pass's parametric amplification:

$$\Gamma_0^2 = (128\pi^3 \omega_1 \omega_2 d_{\text{eff}}^2 / c^3 n_1 n_2 n_3) I .$$

For GaSe crystal in our case  $\Gamma_0^2 [\text{cm}^{-2}] = 1.5 \cdot 10^{-7} I$ , where  $I [\text{W/cm}^2]$ .

For Q-switch regime we take  $I = 20 \text{MW/cm}^2$  which is lower than the damage threshold at single laser pulse (see lower).

So if we take laser intensity  $I = 20 \text{ MW/cm}^2$  we get the increment of parametric amplification

$$\Gamma_0 = 1.76 \text{ cm}^{-1} .$$

The single pass gain  $G(L) = (\Gamma_0 L)^2 = 3$  (for weak gain approximation).

For mode-locking regime with the same focusing

$$\Gamma_0 = 8.8 \text{ cm}^{-1}$$

The single pass gain:  $G(L) = 1/4 \exp(2\Gamma_0 L) = 1.1 \cdot 10^7$  (for large gain approximation).

The obtained value of single pass gain shows us the possibility of obtaining resonator OPO on GaSe pumped by a 3 micron YSGG:Cr:Er laser. In the case of mode-locking regime we shall consider an OPO with synchronous pumping.

Estimation of other parameters of type I and II of phasematching (Table 2):

Table 2.

	Type I	Type II
External angle(at degenerate point), deg	30	44
Parametric amplification linewidth, cm <sup>-1</sup>	400	40
Optical walk-off, rad	$5.46 \cdot 10^{-2}$	$7.53 \cdot 10^{-2}$
mm, for 1 cm crystal length	0.55	0.75
Fresnel losses , %	25	31
o		
e	16(pump)	11
Efficient nonlinearity, d <sub>eff</sub> , cm din <sup>1/2</sup>	$d_{22} \cos\theta \sin 3\phi$	$d_{22} \cos^2 \theta \sin 3\phi$
	$1.28 \cdot 10^{-7}$	$1.21 \cdot 10^{-7}$

The main difference in consideration Type I and II phasematching is approximately 10 times wider amplification linewidth in Type I phasematching. It leads to lower spectral peak gain and thus higher pump threshold. Thus in experimental consideration we shall start with Type II phasematching. However wide spectral linewidth of parametric amplification at the degenerate point might be very helpful for inverse nonlinear process as second harmonic generation of wide spectral signal near 5-6 microns(e.g. CO laser). The pump beam dimension for OPO should be larger than 1 mm in the plane of walk-off to decrease signal and idle wave walk-off from the pump area.

Because of high Fresnel reflection from the faces of the crystal we are going to use both mirrors of OPO resonator with high reflection at resonant waves and coupling through Fresnel losses. The Fresnel losses are higher for ordinary wave, from this point of view the type II phasematching is also more attractive to make resonance of OPO cavity on shorter signal wave and coupling from longer idle wave due to reflection.

Among well studied lasers on ion-doped crystals lasing near 3  $\mu\text{m}$  at room temperature, there are two major materials: YAG:Er( $\lambda=2.94 \mu\text{m}$ ) and YSGG:Cr:Er( $\lambda=2.79 \mu\text{m}$ ). For experiments with mid IR optical parametric oscillator(OPO) we choose YSGG:Cr:Er laser. Yttrium Scandium Gallium Garnet(YSGG:Cr:Er) has better output

parameters at low level pumping due to higher conversion efficiency of the flash lamp pumping, because of Cr in the host material. Spectral amplification bandwidth of YSGG:Cr:Er is large enough for mode locking( $\delta\nu=12\text{ cm}^{-1}$ ) and wider than in YAG:Er.

In the laser system which combines both Q-switching and mode-locking we tested two different approaches. The first is using plane-parallel laser rod with AR coatings and the second- Brewster cut laser rod. The comparison of two set-ups is presented in the following table:

Table 3.

	Brewster cut laser rod	Plane-parallel laser rod with AR
Threshold, free running	20 J	14 J
Threshold, free running, with $\text{LiNbO}_3$	25 J	22 J
Q-switcher intracavity		
Output Energy, $\text{TEM}_{00}$	20-25 mJ	10 mJ( at higher level damage of AR)
Pulse duration	200-230 ns	110-130 ns
Comments on using	<ol style="list-style-type: none"> <li>1. Lower intensity on laser rod faces</li> <li>2. Absence of residual mode selection</li> <li>3. Longer pulse duration is better for OPO: longer build-up time</li> <li>4. More sophisticated adjustment</li> </ol>	<ol style="list-style-type: none"> <li>1. Poor quality of AR coating at <math>3\text{ }\mu\text{m}</math>, as a consequence damage of AR and laser rod faces.</li> <li>2. Residual mode selection</li> <li>3. Shorter pulse duration</li> <li>4. Easier adjustment</li> </ol>

The optical setup of the laser is shown at the Fig.2.

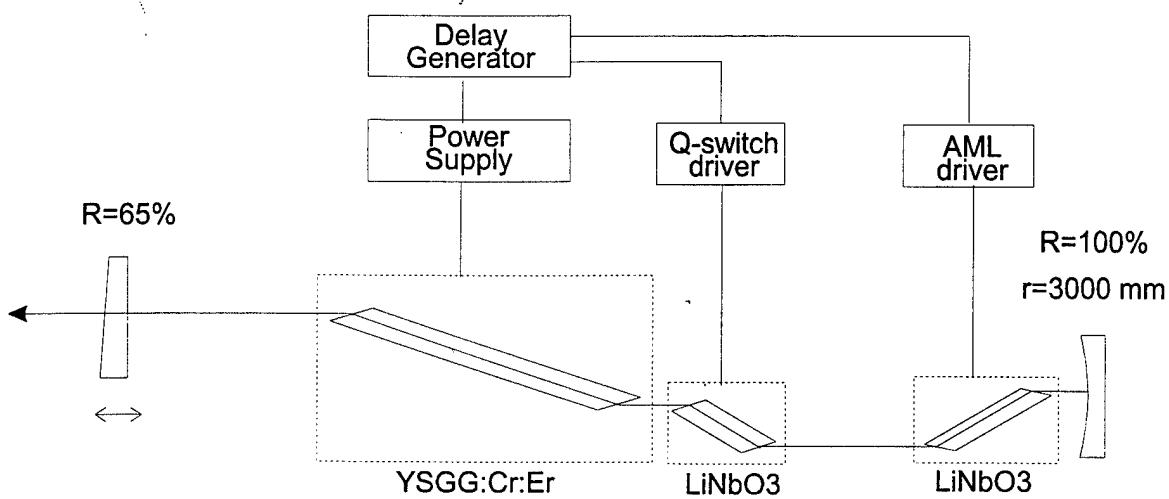


Fig.2

For cavity and HF loss modulation we used  $\text{LiNbO}_3$  Pockels cells.

Parameters of optical elements:

Laser rod: YSGG:Cr:Er of diameter 4x60 mm, supplied by ELMA company.

Electro-optical Q-switcher:  $\text{LiNbO}_3$  6x12x20 mm,

Electro-optical Active Mode-Locker:  $\text{LiNbO}_3$  3x6x36 mm, thinner and longer dimensions were made to increase modulation depth.

Resonator length  $L_{\text{res}}$  (between mirrors) = 113 cm, it corresponds to  $f=120$  MHz inter-mode beating frequency.

Features of electrical part of electro-optical setup of the laser.

Q-switching driver. Independent driver with independent delay. Modulator is "opened" at "zero" applied voltage.  $U_{\text{stat}} = 1-2$  kV,  $U_{\text{pulse}} = 3-4$  kV,  $\tau_{\text{pulse}} = 2 \mu\text{s}$ .

Active mode-locking driver. Quartz stabilized master oscillator,  $f= 60$  MHz,  $\delta f/f = 10^{-5}$ , half of inter-mode beating: modulator is "opened" twice for a period at "zero" points of applied sinus HF voltage.

HF modulator is a "capacitor" in a resonance contour in anode circuit of HF triode bulb. It results in q-factor increasing of HF voltage ( $U= 3-5$  kV) on the modulator.

Features of optical setup of the laser.

Intracavity polarizing was just due to Brewster faces of optical elements. This excluded problem with linear polarizer's in 3  $\mu\text{m}$  band. This optical approach is suitable only for not high single pass gain laser media.

Exact adjustment between HF cavity loss modulation and inter-mode beating was made by accurate tuning of resonator length (accuracy  $\delta L = 10 \mu\text{m}$ ).

Capability of a laser with this optical setup is an independent work in two regimes: pure Q-switching and active mode-locking (with Q-switching). The change between these regimes was by electrical switching.

Measured output parameters of the YSGG:Cr:Er laser with Q-switching and active mode-locking :

repetition rate 1-3 Hz, typical working pump level 45 J, TEM<sub>00</sub> mode,  $w_0 = 0.6 \text{ mm}$

Q-switch:  $E_{\text{out}} = 22 \text{ mJ}$ ,  $\tau = 210 \text{ ns}$

Active mode-locking:  $E_{\text{out}} = 16 \text{ mJ}$ ,  $\tau_{\text{env}} = 210 \text{ ns}$ , 30-35 spikes at HM, estimated single spike pulse duration 100-200 ps.

Output energy might be increased since the power supply had enough energy stored. We did not do that to exclude damage of optical elements in the laser cavity with used output coupler.

We estimated that output energy would be enough for first trials of OPO on GaSe crystal. This was also one of the reasons why we have not tried amplifier for the described master oscillator.

In the first set of experiments with GaSe OPO we studied the type II phase matching since as we reported in our first report in this case we have higher spectral brightness in parametric waves (lower threshold and lower requirements for dielectric mirrors) and parametric waves are different in polarization (easier to obtain single resonator regime).

The experimental set-up OPO with Q-switched pump laser is presented on the Fig.3. The pump laser pulses were Q-switched:  $E_{\text{out}} = 20-22 \text{ mJ}$ , TEM<sub>00</sub>,  $w_0 = 0.6 \text{ mm}$ ,  $\tau = 210 \text{ ns}$ . The pump energy on crystal was 13-14 mJ and the beam waist  $w_0 = 0.6 \text{ mm}$ . GaSe crystal had aperture diameter 11 mm and thickness 8 mm.

Single pass parametric amplification:

$$\Gamma_0^2 [\text{cm}^{-2}] = 1.5 \cdot 10^{-7} I, \quad \text{where } I [\text{W/cm}^2].$$

In our case we have  $I = 4.4 \text{ MW/cm}^2$ ,  $\Gamma_0 = 0.8 \text{ cm}^{-1}$ , the single pass gain  $G(L) = (\Gamma_0 L)^2 = 0.42$  (for weak gain approximation).

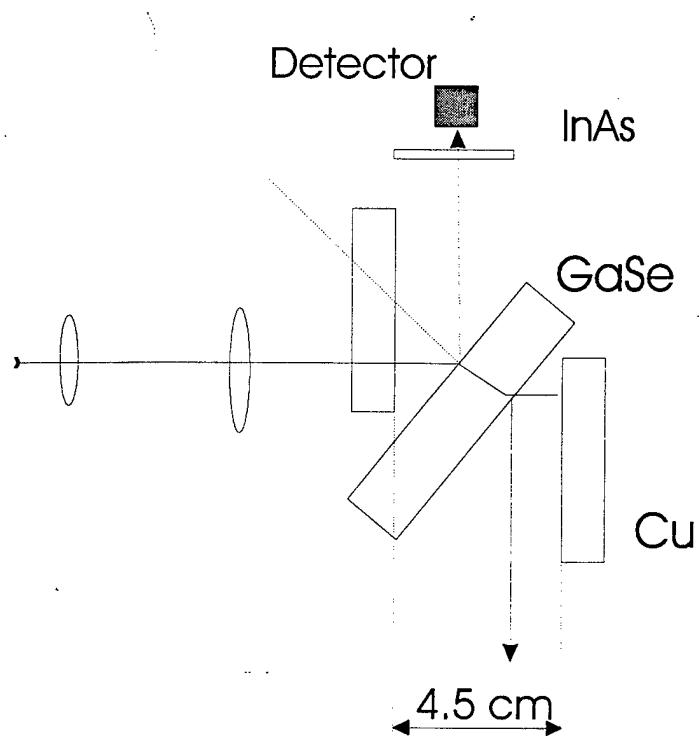


Fig.3

The threshold of parametric oscillation was found at the pump energy  $E_p=11-12$  mJ.

The output coupling of OPO resonator was due to Fresnel losses from the surface of GaSe crystal, we estimated coupling as  $R=0.32$ . We consider this coupling is very high and for future work it is essential to find a way to make AR coatings on the GaSe crystal. Then the coupling of the OPO resonator will be not by Fresnel reflection but through the more reflective output mirror(instead of metal one). This could substantially decrease the threshold and optimize the output coupling of the OPO. The IR signal was detected by Ge-Au liquid nitrogen cooled detector and the energy of IR signal was measured by calibrated pyroelectric detector ( $\eta=0.36$  J/V) on the one of the reflections. Close to the degenerate point the maximum energy in both IR parametric waves was  $E_{IR}=0.45$  mJ. By rotation the crystal at  $J_{ext}=+-1.5^\circ$ , that corresponded to  $J_{int}=+-0.6^\circ$  we obtained tuning of IR signal around  $4.9-6 \mu\text{m}$ (according to the phase matching curve) without adjustment of OPO resonator. This tuning range was limited by reflectance range of the input OPO mirror and misalignment of the OPO resonator.

Using the train of mode-locked picosecond pulses (active mode-locking:  $E_{out}= 16$  mJ,  $\tau_{env}=210$  ns, 30-35 spikes at high frequency modulation, pulse duration 100-200 ps). We obtained and detected signal of parametric superfluorescence for both type I and type II cases. The tuning range was estimated by the angle of rotation of the crystal. For the

type I phasemathing the tuning range was 3.5-14  $\mu\text{m}$ , and for type II - 4-9  $\mu\text{m}$ . The maximum conversion efficiency was about 1%.

In our opinion it is realistic to increase output parameters of GaSe OPO with the following modification:

- using cylindrical lenses in the telescope(optical focusing system) in order to enlarge beam in the plane of the optical walk-off and focusing in the perpendicular direction.
- increase pump intensity.

We found that damage threshold of GaSe crystal by 200 ns laser is  $2\text{J/cm}^2$  (for a few pulses), thus we can increase pump intensity in 2 times. The damage threshold by the train of mode-locked picosecond pulses is about the same value that says that pulses in the train are additive for the damage.

The best approach to increase pump power is to use a new laser crystal recently obtained at GPI is YSGG:Cr:Yb:Ho( $\lambda=2.92 \mu\text{m}$ ), that have similar lasing parameters as YSGG:Cr:Er( $\lambda=2.79 \mu\text{m}$ ), but higher single pass amplification gain. In this case would be possible to increase diameter of laser beam and then to increase the length of parametric interaction and use more thick samples of GaSe crystals.

Our first experience in working with GaSe OPO pumped by Q-switched pulses shows that pumping of GaSe by 2  $\mu\text{m}$  laser radiation (a holmium laser) is also possible, while the tuning range in this case is better fit to the window of transparency of atmosphere(3.5-4.5  $\mu\text{m}$ ).

Returning to estimations:

Single pass parametric amplification:  $G = (\Gamma_0/\Gamma)^2 \text{sh}^2(\Gamma L) - \delta \times L$ ,

where  $\Gamma^2 = \Gamma_0^2 - (\Delta k/2)^2$ ,  $\Delta k$  - wave mismatching,  $\delta[\text{cm}^{-1}]$  - week intracavity losses,  $L$  - length of parametric interaction and

$$\Gamma_0^2 = (128\pi^3 \omega_1 \omega_2 d_{\text{eff}}^2 / c^3 n_1 n_2 n_3) I .$$

For GaSe crystal in our case  $\Gamma_0^2 [\text{cm}^{-2}] = 1.5 \cdot 10^{-7} I$ , where  $I [\text{W/cm}^2]$

For Q-switch regime we take  $I = 20\text{MW/cm}^2$  which is lower than damage threshold. The actual damage threshold of GaSe crystal by 3 micron laser radiation was roughly determined in this work and is close to  $2 \text{ J/cm}^2$ .

In GaSe crystal we have  $\Gamma_0 = 1.73 \text{ cm}^{-1}$ .

The single pass gain for  $L=1\text{cm}$   $G(L) = (\Gamma_0 L)^2 = 3$  (for weak gain approximation).

Fresnel losses gives large enough equivalent intracavity losses  $\delta[\text{cm}^{-1}]$  apart from losses inside GaSe crystal. This is a reason of high pump threshold of OPO on GaSe. The obtained value of single pass gain shows us the possibility of obtaining resonator OPO on GaSe pumped by a 3 micron YSGG:Cr:Er laser but very close to the threshold of the laser-damage.

Estimation of other parameters of type I and II of phasematching:

Table 4

		Type I	Type II
External angle(at degenerate point), deg		30	44
Parametric amplification linewidth, $\text{cm}^{-1}$		400	40
Optical walk-off, rad		$5.46 \cdot 10^{-2}$	$7.53 \cdot 10^{-2}$
	mm, for 1 cm crystal length	0.55	0.75
Fresnel losses , %	o	25	32
	e	16(pump)	11
Efficient nonlinearity, $d_{\text{eff}}$ , $\text{cm din}^{1/2}$		$d_{22}\cos\theta \sin 3\phi$ $1.28 \cdot 10^{-7}$	$d_{22}\cos^2\theta \sin 3\phi$ $1.21 \cdot 10^{-7}$

#### Important remark

As we occur during the work we must take into account humidity because of its influence on intracavity losses and on laser damage of optical elements. As we measured, absorption coefficient for the room atmosphere  $\alpha(\lambda=2.79 \mu\text{m}) \approx 4 \times 10^{-3} \text{ cm}^{-1}$ . It means that laser intensity decreases by e-times on the distance  $L(1/e)=250 \text{ cm}$ . This distance is very close to distance of round trip in resonator of our Er-laser. Of course, the absorption coefficient  $\alpha(\lambda=2.79 \mu\text{m})$  depends on the value of humidity and this dependence must be search in course of some other work.

#### Laser damage of GaSe crystal

The actual damage threshold of GaSe crystal by 3 micron laser radiation in Q-switched mode was roughly estimated in this work for single laser pulse and depends on the crystal's sample, position(location) of irradiation and humidity. Dependence on humidity has very simple explanation: high absorption coefficient of the water at  $\lambda=2.79 \mu\text{m}$  – wavelength of the Er-laser.

To measure damage of GaSe crystal by Er-laser radiation we had fist to measure parameters of laser's beam ( $\text{TEM}_{00}$ -mode), i.e. parameters of gaussian beam. We used for that two pin-holes of diameters 1.07 mm and 0.8 mm. The pin-hole method gives the expression for the radius of gaussian beam  $w$ :

$$w(z) = (R\sqrt{2})/\{-\ln[1 - (E(R)/E(\infty))]\}^{1/2},$$

where  $R$  - radius of pin-hole,  $E(\infty)$  - whole energy of gaussian beam,  $E(R)$  - beam's energy which gone through the pin-hole centered with axis  $z$  of gaussian beam,  $z$  - the distanse between pin-hole and waist  $w_0 = w(0)$ . By means of measuring  $w(z_1)$  and  $w(z_2)$  and using formula

$$w^2(z) = w_0^2 + (\lambda/\pi)^2(z^2/w_0^2),$$

we can get  $w_0$  also. We got in result with pin-hole  $D_1=1.07$  mm -  $w_1(104\text{cm}) = 0.23$  cm,  $w_1(38\text{cm}) = 0.13$  cm, with pin-hole  $D_2=0.8$  mm -  $w_2(104\text{cm}) = 0.29$  cm,  $w_2(38\text{cm}) = 0.19$  cm, and  $w_{01} = 0.045$  cm,  $w_{02} = 0.039$  cm. We don't know exactly the position of beam's waist because of thermolens in our laser's rod. Then we assumed that the waist is located on the flat output mirror of laser's resonator and we used  $w_0 = (w_{01}+w_{02})/2=0.042$  cm. At this assumption we have dependence of radius  $w(z)$  on distance  $z$  from the waist  $w_0$ , shown on the next figure (Fig.4):

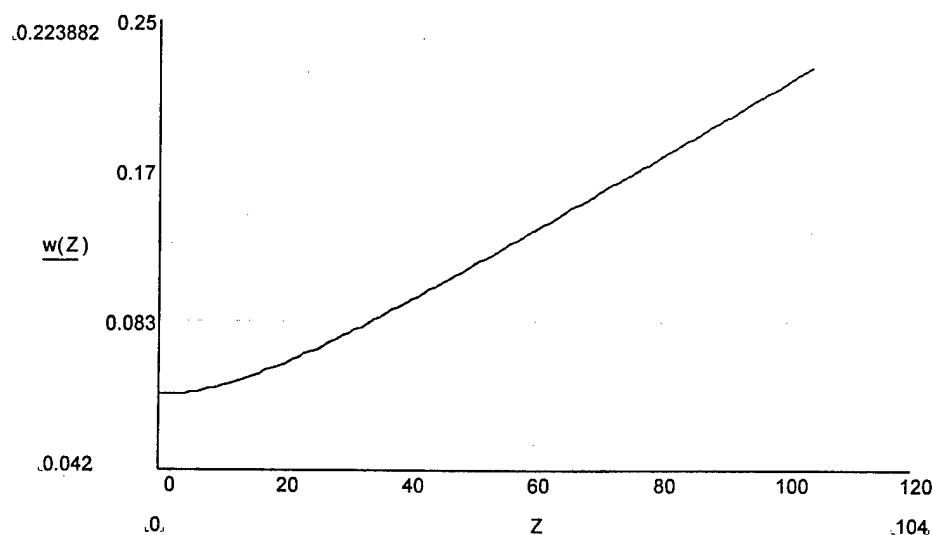


Fig.4

We installed  $\text{BaF}_2$  focusing lens with focal length  $f=14.5 \text{ cm}$  on the distance  $z=104 \text{ cm}$  from the flat output mirror (as the waist  $w_0$  location). The focusing lens was located in the far zone of the gaussian beam as it is seen on the Fig.5.

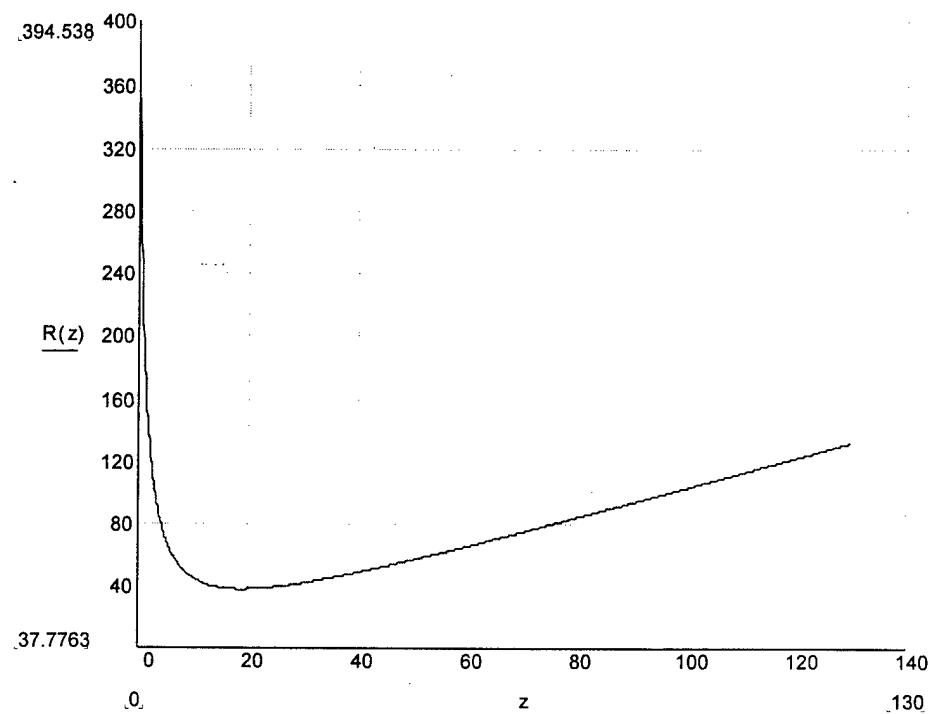


Fig.5

The dependence of gaussian beam's radius  $w(z)$  on distance  $z$  from the  $\text{BaF}_2$  lens is shown on the Fig.6.

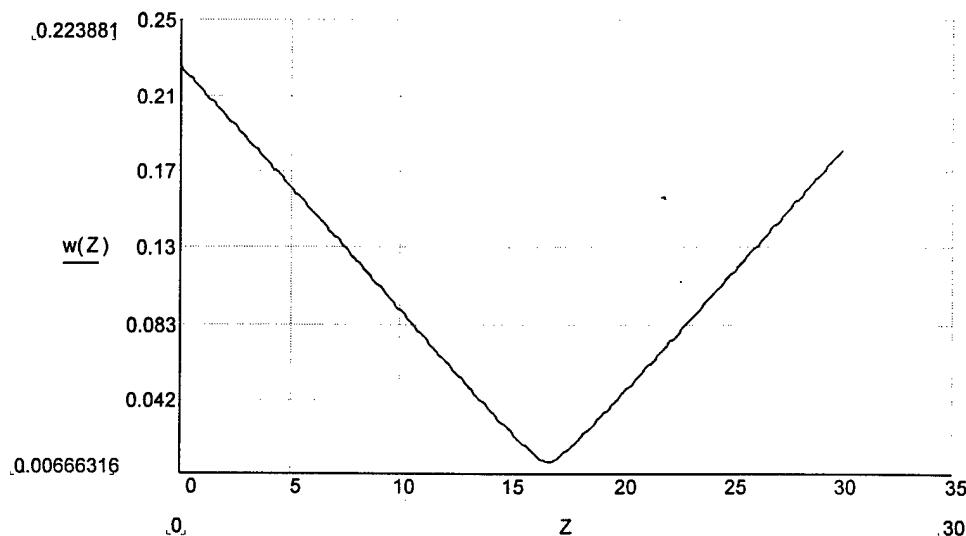


Fig.6

The laser's gaussian beam behind the  $\text{BaF}_2$  lens has the waist location near  $z = 14$  cm; we placed a sample of GaSe crystal far enough from the lens and moved this sample toward the lens until damage could be seen. We occurred the damage after many ( $>10\ldots20$ ) laser's shots at the distance  $z = 21.5$  cm from the lens. The energy of laser's pulse on the lens was equal  $E = 11$  mJ. The damage after single laser pulse was occurred at the distance  $z = 19$  cm. Thus we have  $E = 11$  mJ which is the energy of Er-laser radiation on the surface of GaSe crystal. The spot size  $S(z) = \pi w^2(z)/2$  and  $F(z) = E/S(z)$  is energy's density in  $\text{J/cm}^2$ . Damage of GaSe irradiated by many ( $>10\ldots20$ ) laser pulses at  $F = 1.7 \text{ J/cm}^2$ . Damage of GaSe at one laser pulse occurs at  $\Phi = 7 \text{ J/cm}^2$ .

We were planning to conclude experiments on synchronous pumping by the train of picosecond mode locked pulses of GaSe OPO resonator with optical length equal to that of pump laser, but we were stopped by difficulties connected with high enough absorption of the laser radiation ( $2.79 \mu\text{m}$ ) in the air.

We were planning specially to consider theoretically and experimentally the phasematching condition when  $\omega_s = 2\omega_i$ , or  $\omega_p = 3\omega_i$ , and the IR resonator is matched for  $\omega_s$ . In our case of pump by  $2.79 \mu\text{m}$  radiation,  $\omega_s = 4.19 \mu\text{m}$  lays in the transparency window of the atmosphere. It is expected higher conversion efficiency in this case when

signal and idle waves of OPO are in second order nonlinear resonance. But we hadn't enough time to do that.

## SUMMARY

This research demonstrate an optical parametric oscillation(OPO) in a GaSe crystal pumped by an actively Q-switched and mode-locked three micron erbium laser. Both type I and II of phasematching condition experimentally considered.

Resonant optical parametric oscillation in GaSe crystal has been demonstrated for the first time with pump by a Q-switched YSGGCr:Er( $\lambda=2.79 \mu\text{m}$ ) laser. Single resonator type II (e-oe) OPO was obtained near the degenerate point. Tunability around  $4.9\text{-}6 \mu\text{m}$  was observed without adjustment of OPO resonator. The highest output conversion to both parametric waves was measured to be 7%. Parametric superfluorescence for both type I and II phase matching was obtained by pumping GaSe by a train of mode-locked picosecond pulses, IR signal was tuned up to  $14 \mu\text{m}$ . Maximum conversion efficiency in this case was about 1%.

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